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DETERMINATION OF COUNTING EFFICIENCY OF ALPHA-COUNTERS WITH RADON AND THORON DAUGHTER SOURCES

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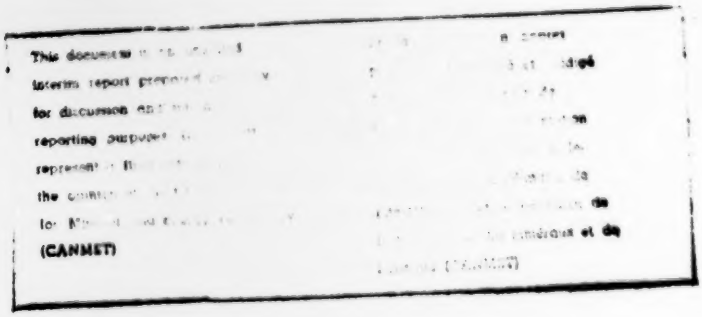
SEPTEMBER 1982



17 pp

MINERALS RESEARCH PROGRAM
MINING RESEARCH LABORATORIES
DIVISION REPORT MRP/MRL 82-84(TR)

MICROMEDIA

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by

J. Bigu* and G.K. Szlapetis**

ABSTRACT

Measurements of α-counting efficiency of an α-counter were carried out using two recently developed radon daughter and thoron daughter sources manufactured by Pylon Electronics and known commercially as radon daughter standard Rn-190 and thoron daughter standard Th-190, respectively. The α-counting efficiency of the α-counter was also determined with standard Am-241 sources supplied by the Ontario Ministry of Labour. Some disagreement was observed between the values obtained for the counter efficiency using the Am-241, Rn-190 and Th-190 sources. The possible reasons for this disagreement are briefly indicated.

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INTRODUCTION

Uranium (U-238) and thorium (Th-232) are the parents of two natural radioactive chains. They are distributed more or less uniformly in the crust of the Earth. Through a series of radioactive decays U-238 and Th-232 become Ra-226 and Th-228, respectively. Ra-226 and Th-228 decay, respectively, into radon gas (Rn-222) and thoron gas (Rn-220). Rn-222 in turn decays into the so-called short-lived radon daughters RaA, RaB, RaC and RaC', whereas Rn-220 decays into its short-lived thoron daughters ThA, ThB, ThC, ThC' and ThC", (see Appendices 1 and 2).

Because the radon and thoron progenies pose a potential health hazard to occupational workers in the uranium industry, their airborne levels must be carefully monitored. Although alpha (α), beta (β) and gamma (γ) radiation is emitted by the decay of different decay products, i.e., daughters, only α-radiation needs to concern us here. Alpha radiation can be determined by a wide range of commercially available instrumentation. However, reliable measurement of α-radiation requires accurate determination of the instruments' α-counting efficiency, say ε, with reference standard radioactive α-sources, e.g., Am-241, Cm-244 and Pu-239.

Most radioactive sources used for determining the α-counting efficiency of α-detectors and associated electronic circuitry emit α-particles of lower energy than those corresponding to the radon progeny (6 MeV for RaA and 7.7 MeV for RaC') and thoron progeny (6.1 MeV for ThC and 8.8 MeV for ThC'). For example the approximate α-energies corresponding to Am-241, Cm-244 and Pu-239 are 5.5 MeV, 5.8 MeV and 5.1 MeV, respectively. Because α-counting efficiency for a given geometry might be energy-dependent for certain α-detectors, it is of interest to determine the efficiency at the desired α-energy, preferentially with standard sources of the same radioisotopes of interest.

Pylon Instruments (Ottawa) has recently developed radon daughter and thoron daughter standard sources for α-energy calibration and α-counting efficiency determination of α-radiation instrumentation. They are commercially available under the name Radon Daughter Standard Rn-190 and Thoron Daughter Standard Th-190.

This report presents data pertaining to the use of the above standard sources for the determination of the α-counting efficiency of α-counters. The data obtained was compared with that obtained with several Am-241 standard sources supplied by the Ontario Ministry of Labour. The Rn-190 and Th-190 standard sources were also used in conjunction with a technical evaluation of an automated radon daughter grab-sampler manufactured by Pylon Instruments under the commercial name WL-1000C (1,2).

RADIOACTIVE SOURCES

The two calibration standard sources Rn-190 and Th-190 used in these tests are sealed inorganic salts of Ra-226 and Th-228, respectively. Each source was prepared from a weighed aliquot of a solution whose specific activity was determined by the National Bureau of Standards with an estimated error of about 1.2% (1,2).

The source holder is made up of an aluminum chamber, with the source embedded on the inner top side, and a base plate. A rubber gasket is placed between the chamber and base plate which are clamped together to make the system gas leak free. A filter is placed in a recess on the base plate completing the radioactive standard source system. Ra-226 and Th-228 decay, respectively, into Rn-222 and Rn-220, thereby filling the chamber. The radioactive gases decay in turn into their short-lived decay products which attach themselves to the walls of the chamber (i.e., plate-out) including the

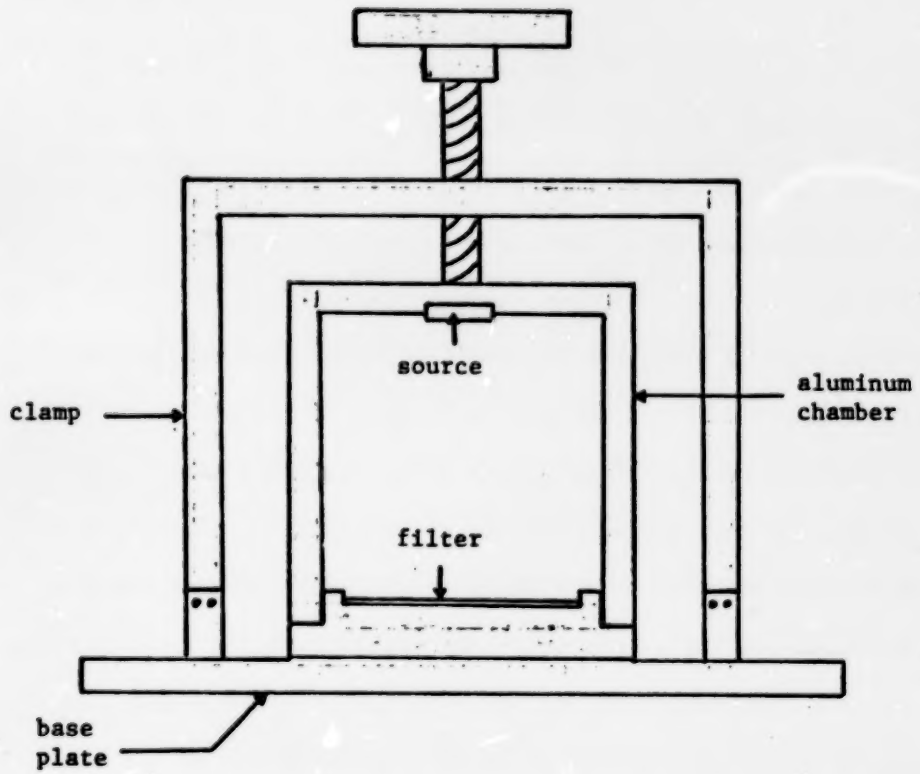

Fig. 1 - Standard source used in this experiment. Source is an inorganic salt of either Ra-226 or Th-228. Filter is 0.8 μm Millipore.

filter. It is claimed that the attachment is uniform, independent of the kind of surface, including the filter material, and of environmental factors. The activity deposited on the filter is proportional to the ingrowth time, i.e., time the filter remains in the chamber, and the strength of the Ra-226 or Th-228 source. The filter activity is used to determine the α-counting efficiency of α-counters and other instrumentation.

EXPERIMENTAL PROCEDURE

The standard sources Rn-190 and Th-190 were used to determine the α-counting efficiency of the WL-1000C and an α-scaler manufactured by Tri-Mac (Winnipeg) under the commercial name TM-372A (3). The results of the former have been given elsewhere (4). This report describes work done in conjunction with the TM-372A.

Type AA 0.8 μm Millipore filters are placed into the Rn-190 and Th-190 source chambers for a given period of time to collect activity. This time period will be referred to hereafter as the ingrowth time. The filter is then removed and its activity measured by the TM-372A.

Alpha-counting was carried out for different counting periods according to Pylon analytical procedures as indicated below and described elsewhere (1,2). Three counting periods were used: a 5-min and two 15-min taken, respectively, 2-min, 8-min and 24-min after the end of the ingrowth period. The total α-activity measured during these three counting periods will be denoted hereafter as I(2-7), I(8-23) and I(24-39), respectively.

Measurements were conducted for ingrowth times ranging from 1 h to 240.5 h. Temperature, relative humidity and barometric pressure at the time the filter was placed in (and removed from) the chamber were noted to see if these environmental factors had any effect on the deposition of the short-lived

radon and thoron progenies on the filter.

THEORETICAL BACKGROUND

The α-counting efficiency, ε, of a detector is defined by:

$$\varepsilon = (\text{cpm/dpm}) \times 10^2 \qquad \text{Eq 1}$$

Where cpm represents the α-count rate in counts per minute obtained from the instrument, e.g., TM-372A, and dpm is the disintegration rate, in disintegrations per minute, of the standard calibration source used.

The α-counting efficiency can be determined using Pylon's procedure as follows:

$$\varepsilon = \frac{I(t_1 \rightarrow t_2)}{A \times F \times Y} \times 10^2 \qquad \text{Eq 2}$$

where, $I(t_1 \rightarrow t_2)$ is the count for one of the counting periods I(2-7), I(8-23) or I(24-39), with t_1 being the time elapsed since the opening of the source and t_2 being the end of the counting period.

A is the activity of daughters in equilibrium with ingrown radon or thoron. The activity A is given by Equation 3:

$$A = \text{source strength (dpm)} \times (1 - \text{decay factor}). \qquad \text{Eq 3}$$

The term in brackets represents the fraction of Rn-222 or ThB accumulated from the decay of Ra-226 or Th-228, respectively. The decay factor is simply $e^{-\lambda x t}$ where λ is the radioactive decay constant of x, where x is either Rn-222 or ThB. Since $\lambda = 0.693/T_{\frac{1}{2}}$, where $T_{\frac{1}{2}}$ is the half-life of x, it is easy to show that λ (Rn-222) $= 1.26 \times 10^{-4}$ min^{-1} and $\lambda_{ThB} = 1.09 \times 10^{-3}$ min^{-1}.

The source strength in Equation 3, say $A_s(t)$, represents the activity of Ra-226 or Th-228, in dpm, at the time the ingrowth period begins. If A_s° is the activity of Ra-226 or Th-228 at the time the sources are first calibrated,

$$A_s(t) = A_s^{\circ} e^{-\lambda x t} \qquad \text{Eq 4}$$

(Note: A_s° is usually given in pCi. In order to convert pCi to dpm multiply by 2.22, i.e., 1 pCi = $10^{-12} \frac{Ci}{pCi} \times 3.70 \times 10^{10} \frac{dps}{Ci} \times \frac{60}{1} \frac{s}{min}$ = 2.22 dpm).

F is a correction factor for the counting periods chosen. The values of F as given by Pylon are as follows (1,2):

For Rn-190

F = 6.891 for the count period I(2-7)

F = 14.907 for the count period I(8-23)

F = 12.05 for the count period I(24-39)

For Th-190

F = 4.991 for the count period I(2-7)

F = 14.950 for the count period I(8-23)

F = 14.883 for the count period I(24.39)

Y is defined as the ratio of the filter surface area to the total chamber surface area. The value given by Pylon is: Y = 0.123.

RESULTS AND DISCUSSION

The results obtained in this experiment are summarized in Tables 1 and 2. The counting efficiency was calculated for each counting period and the three efficiencies were averaged. Included in the Tables are the α-counts with which the efficiencies were calculated.

The average percent counting efficiency was plotted vs time for the Rn-190 (Fig. 2) and Th-190 (Fig. 3) standards. When there were more than one efficiency value for a given period of time their average was calculated and plotted.

Relative humidity, temperature and barometric pressure taken at the time the filters were placed in the source chamber show no correlation between

Table 1 - Percent counting efficiency of the Sample Counter TM-372A as determined by the Rn-190 source for different counting periods and ingrowth time.

Growth Time,h	I(2-7) α-count	I(2-7) ε(%)	I(8-23) α-count	I(8-23) ε (%)	I(24-39) α-count	I(24-39) ε (%)	Average % (ε) Counting Efficiency
1	758	21.23	1450	18.77	1303	20.85	20.28
	624	17.48	1153	14.92	975	15.60	16.00
2	1618	22.97	3329	21.82	2376	19.26	21.35
3	2422	23.01	5138	22.56	4306	23.38	22.98
	2470	23.47	5289	23.22	4312	23.41	23.36
3.5	2993	24.39	6403	24.13	5249	24.45	24.33
4	3555	25.38	7535	24.86	6231	25.43	25.22
	3602	25.26	7575	25.00	6361	25.96	25.41
5	4697	26.94	10265	27.21	8683	28.46	27.54
	4878	27.98	10531	27.91	8747	28.67	28.19
5.5	5452	28.50	11411	27.57	9496	28.37	28.15
16	16325	30.52	34256	29.60	28381	30.32	30.15
20	19888	30.18	43133	30.26	34854	30.23	30.22
21	20153	29.23	43900	29.43	35687	29.58	29.41
22	22488	31.25	48670	31.26	39846	31.65	31.39
23	23752	31.68	51556	31.78	41780	31.85	31.77
41.5	40992	32.42	90507	33.09	72790	32.90	32.80
64	59439	33.00	129271	33.16	104569	33.17	33.11
65	55673	30.53	120911	30.65	97943	30.70	30.63
120	85427	30.44	185446	30.54	150683	30.70	30.56

Note: The α-count for each of the three counting periods is given with corresponding counting efficiency. The average counting efficiency of the three counting periods for a given ingrowth period is also shown.

Table 2 - Percent counting efficiency of the Sample Counter TM-372A as determined by the Th-190 source for different counting periods and ingrowth time.

Growth Time	I(2-7) Count	I(2-7) % ε	I(8-23) Count	I(8-23) % ε	I(24-39) Count	I(24-39) % ε	Average % Counting Efficiency	Source Activity in pCi used in calculation of counting efficiency
1	1139	18.50	4484	24.32	5367	29.24	24.02	71341
1.5	2390	26.44	7966	29.43	9436	35.02	30.30	70985
2	3430	28.80	11824	33.15	13402	37.74	33.23	71270
	3834	32.39	12821	36.16	14515	41.12	36.56	70844
3	6395	36.90	20660	39.81	22662	43.86	40.19	71341
	6924	40.40	22695	44.21	24525	47.99	44.20	70561
3.5	7831	39.40	25091	42.15	26950	45.48	42.34	71270
4	9592	42.00	30337	44.36	32317	47.47	44.61	71484
4.5	11886	48.24	37012	50.16	39131	53.27	50.56	70915
5	12603	47.03	39085	48.70	40831	51.11	48.95	70491
5.25	13010	45.98	41032	48.42	42686	50.60	48.33	71412
17	37644	58.04	113850	58.61	113817	58.86	58.50	70915
18	38657	58.20	117307	58.98	117908	59.55	58.91	70420
20.5	44274	62.35	132773	62.43	133627	63.12	62.63	70561
22	43677	59.18	131420	59.45	131940	59.96	59.53	70985
28	49266	61.12	147244	60.99	147370	61.32	61.14	70420
40.5	58959	65.70	176903	65.82	176291	65.89	65.80	70844
65	62040	65.43	186306	65.61	185419	65.59	65.54	70561
66	58350	61.05	175089	61.17	174659	61.30	61.17	71056
118	56516	62.52	168666	62.29	168251	62.24	62.35	66356*
240.58	55716	61.18	166850	61.17	166914	61.46	61.27	66822*

Note: The α-count for each of the three counting periods is given with corresponding counting efficiency. The average counting efficiency for a specific ingrowth period is shown as well as the source strength of the Th-190 source used in the calculations.

* Because of the relatively short half-life of Th-228 (i.e., 1.9 y), the source activity was calculated for each day of the ingrowth period and an average was determined and used in the calculations.

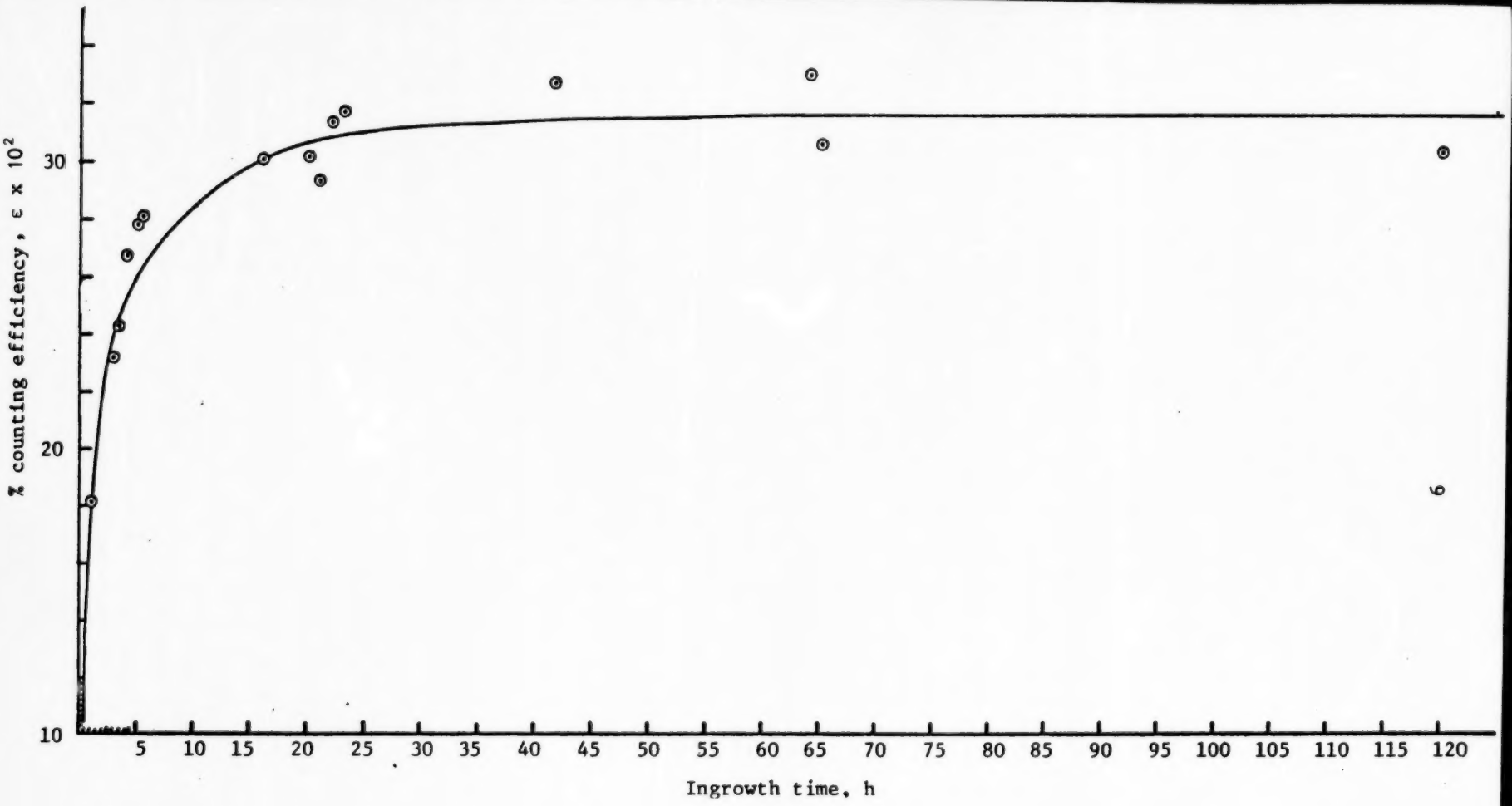

Fig. 2 - % counting efficiency of sample counter TM-372A as determined with the Rn-190 versus ingrowth time.

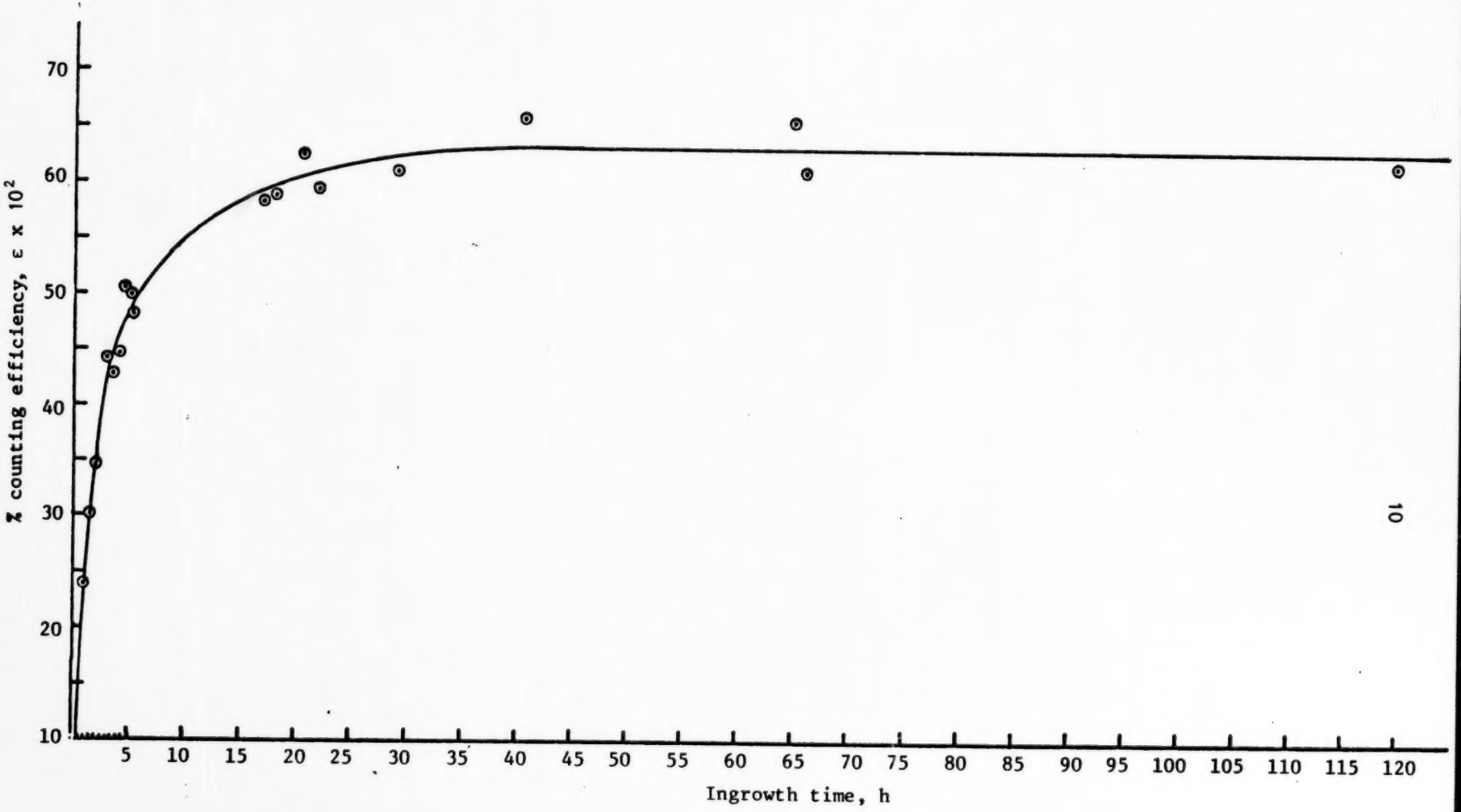

Fig. 3 - % counting efficiency of sample counter TM-372A as determined with the Th-190 versus ingrowth time.

these factors and the counting efficiency.

The counting efficiency follows a power curve for short ingrowth periods (1 to 24 h) and then becomes almost constant with time (Fig. 2 and 3). Therefore, in order to get reliable results, the ingrowth periods must be over 24 h.

The approximate maximum for the counting efficiency of the TM-372A determined using the Rn-190 was 31.99± 1.01%. This same instrument was calibrated for counting efficiency using Am-241 Standard Sources supplied by the Ontario Ministry of Labour. The efficiency determined with Am-241 was 39.99%. This efficiency is 25% higher than the efficiency obtained with the Rn-190 source. Theoretically, the two efficiencies should be approximately the same. In fact, the efficiency determined with Am-241 might be slightly lower because Am-241 emits lower energy alpha-particles than radon daughters. In the case of the Th-190 source the maximum counting efficiency obtained was approximately 63±2.20% which is substantially higher than the theoretical maximum of 50% for 2π geometry.

Independent tests have shown ε, as determined with our Am-241 sources to be correct, the discrepancy observed for ε using the Rn-190 and Th-190 sources, and the Am-241 sources can only be explained in terms of an error in the source strength determination or in the analytical procedure according to references 1 and 2. The authors have recently been informed by Pylon's technical staff (5) that a modification in their operational procedure for the calibration of the radioactive sources renders more reliable values for the sources strength than the technique used previously. A new series of measurements will be conducted when accurate values for the sources strength become available.

ACKNOWLEDGEMENTS

We would like to thank Dr. G. Vandrish for helpful suggestions and his interest throughout this work. We also acknowledge useful discussions with M. Grenier.

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2. Pylon Instruction Manual for Thoron Daughter Standard Source Th-190; 1981.

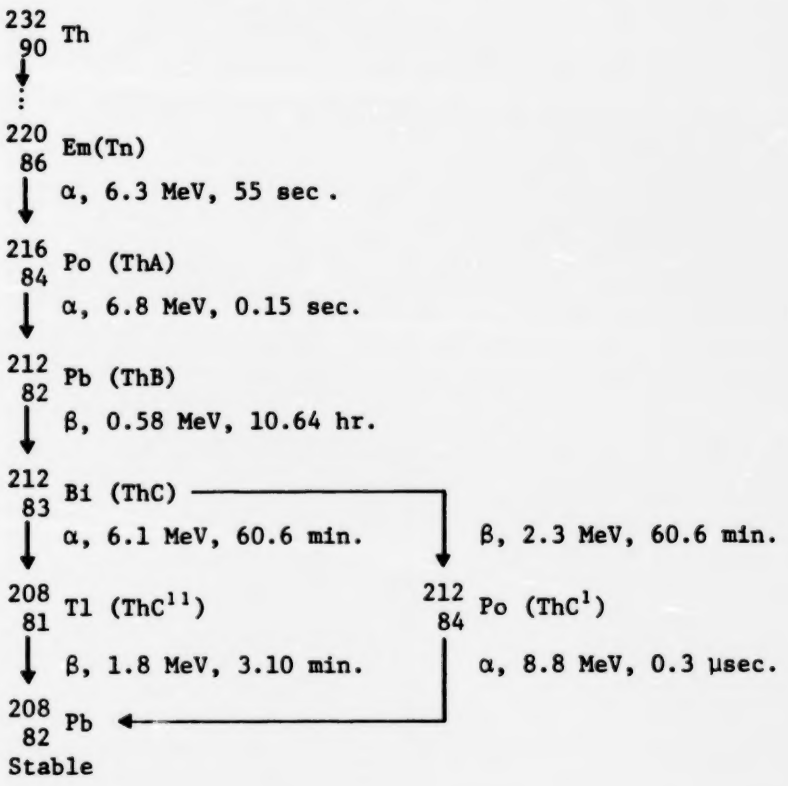
3. Instruction Manual for Model TM-372A Sample Counter: Tri-Met Instruments Ltd., Winnipeg; 1977.

4. Grenier, M. and Bigu, J., "Experimental procedure for the determination of counting efficiency and sampling flow-rate of a grab-sampling Working Level Meter"; Division Report MRP/MRL 82-69 (TR); CANMET, Energy, Mines and Resources Canada; 1982.

5. Vandrish, G., private communication; 1982.

APPENDIX 1

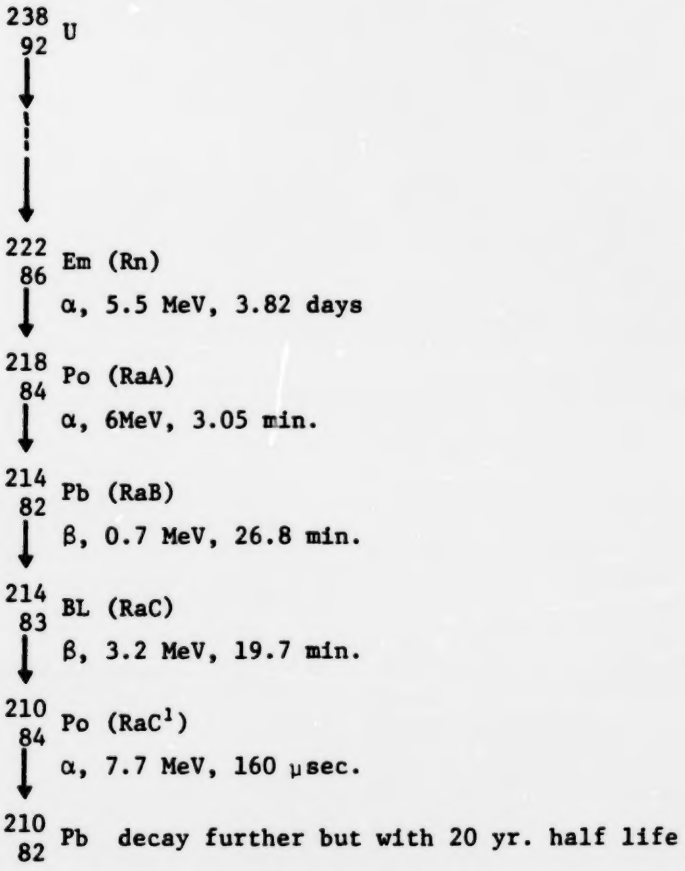
Thorium Series


232 90 Th
220 86 Em(Tn)
α, 6.3 MeV, 55 sec.
216 84 Po (ThA)
α, 6.8 MeV, 0.15 sec.
212 82 Pb (ThB)
β, 0.58 MeV, 10.64 hr.
212 83 Bi (ThC)
α, 6.1 MeV, 60.6 min.
β, 2.3 MeV, 60.6 min.
208 81 Tl (ThC'')
β, 1.8 MeV, 3.10 min.
212 84 Po (ThC')
α, 8.8 MeV, 0.3 μsec.
208 82 Pb
Stable

Decay of thoron gas (Rn-220). Types and energies of the principal emissions and half-lives are shown.

APPENDIX 2

Uranium Series


238 92 U
222 86 Em (Rn)
α, 5.5 MeV, 3.82 days
218 84 Po (RaA)
α, 6MeV, 3.05 min.
214 82 Pb (RaB)
β, 0.7 MeV, 26.8 min.
214 83 BL (RaC)
β, 3.2 MeV, 19.7 min.
210 84 Po (RaC1)
α, 7.7 MeV, 160 μsec.
210 82 Pb decay further but with 20 yr. half life

Decay of radon gas (Rn-222). Types and energies of the principal emissions and half lives are shown.